A Closed Loop Process for the End-of-Life Electric Vehicle Li-ion Batteries: Phase II

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Organization: Worcester Polytechnic Institute

2020 DOE Annual Merit Review

Date: June 1~4, 2020

Project ID #: bat293

Overview

Timeline

- Project start date: Mar. 26, 2018
- Project end date: Sep. 25, 2020
- Percent complete: 90%

Barriers

- Barriers addressed
  - Cost
  - Performance
  - Supply
  - Sustainability

Budget

- Total project funding: $1,083,616
  - DOE share: $541,808
  - Contractor share: $541,808
- Funding received in FY 2019: $593,506

Partners

- Interactions/ collaborations:
  - A123 Systems, Battery Resourcers, Argonne National Laboratory, General Motors, Ford, FCA, SNT
- Project lead: WPI

Relevance and Project Objectives

- During the course of the Phase I USABC program which focused on using recycled batteries to produce NMC111, the team has observed that the xEV battery industry is moving to higher nickel NMCs.

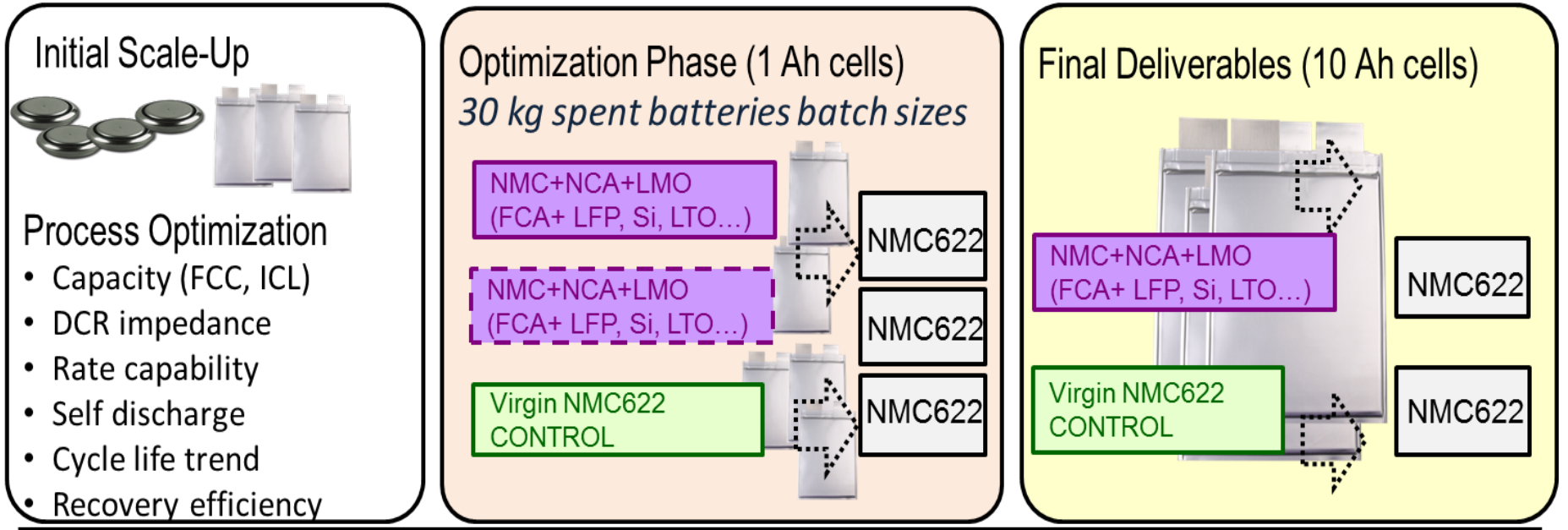
- Building on the successful Phase I program, the overall objective of the Phase II program is to demonstrate the recovery of NMC622 cathode materials from recycled lithium ion batteries with mixed cathode and anode chemistry, and added complexity of adhesives, silicon and LTO that are anticipated materials in the future waste stream.

- The cost model developed for NMC111 will be updated based on the new chemistry process update and scale-up.

Phase II: Milestones

Tasks and Milestones	# of Cells	Timeline (months)- 2 Year Duration								3 month NCE
		Q1, Yr1	Q2, Yr1	Q3, Yr1	Q4, Yr1	Q1, Yr2	Q2, Yr2	Q3, Yr2	Q4, Yr2	Q1, Yr3
Task 1:Optimize the electrochemical properties of recycled NMC622 cathode materials (WPI+BRs)										
1.1 Optimize NMC622 synthesis parameters										
pH, material adding rate, temperature, etc are determined.										
Milestone 1.1: Recovered NMC622 shows similar properties to commercial NMC622										
1.2 Understand the impacts of new anode materials and possible adhesive										
Milestone 1.2:Determine the impacts of anode materials and adhesive										
1.3 Electrodes Development										
Cathode Evaluation: Coin Cell, Full-Cell										
Milestone 1.3: Half cell testing shows properties within 5% of virgin material										
100 coin cells fabricated to evaluate recycled NMC622	100									
Task 1*: NMC622 Coating Development										
Milestone 1*: Coated NMC622 shows similar performance to the commercial coated NMC622										
Task 2: Scale Process to 30 kg batch size (WPI+BRs)										
Milestone 2.1: Scaled process cathode powder within 5% of control										
Deliverable: WPI deliver 1 or 2 batches of 6kg recovered NMC622 to A123 for 1Ah cells										
Deliverable: WPI deliver 15kg recovered NMC622 to A123 for 10Ah cells										
Task 3: Produce 60 1Ah cells from recycled material (1 or 2 input stream+control) (A123)										
Cell Assembly and Formation										
Performance Testing										
Cycle life										
Calendar Testing										
Internal Screening										
Milestone 3.1 Recovered cathode cells within 5% of control cells										
Deliverable: 40 or 60 1Ah cells to USABC/A123	40 or 60									
Task 4: Fabricate large format prismatic 10 Ah cells (1 input stream+control) (A123)										
Internal Screening										
Milestone 4.1 Recovered cathode cells within 5% of control										
Deliverable: 40 10Ah cells to USABC/A123	40									
Task 5: Recycling process update (BRs)										
Milestone 5.1 Verify that the recycling process is economical feasible										
Deliverable: Updated recycling process and cost analysis										
Final Report (WPI)										

Phase II: Cell Fabrication and Test Plan

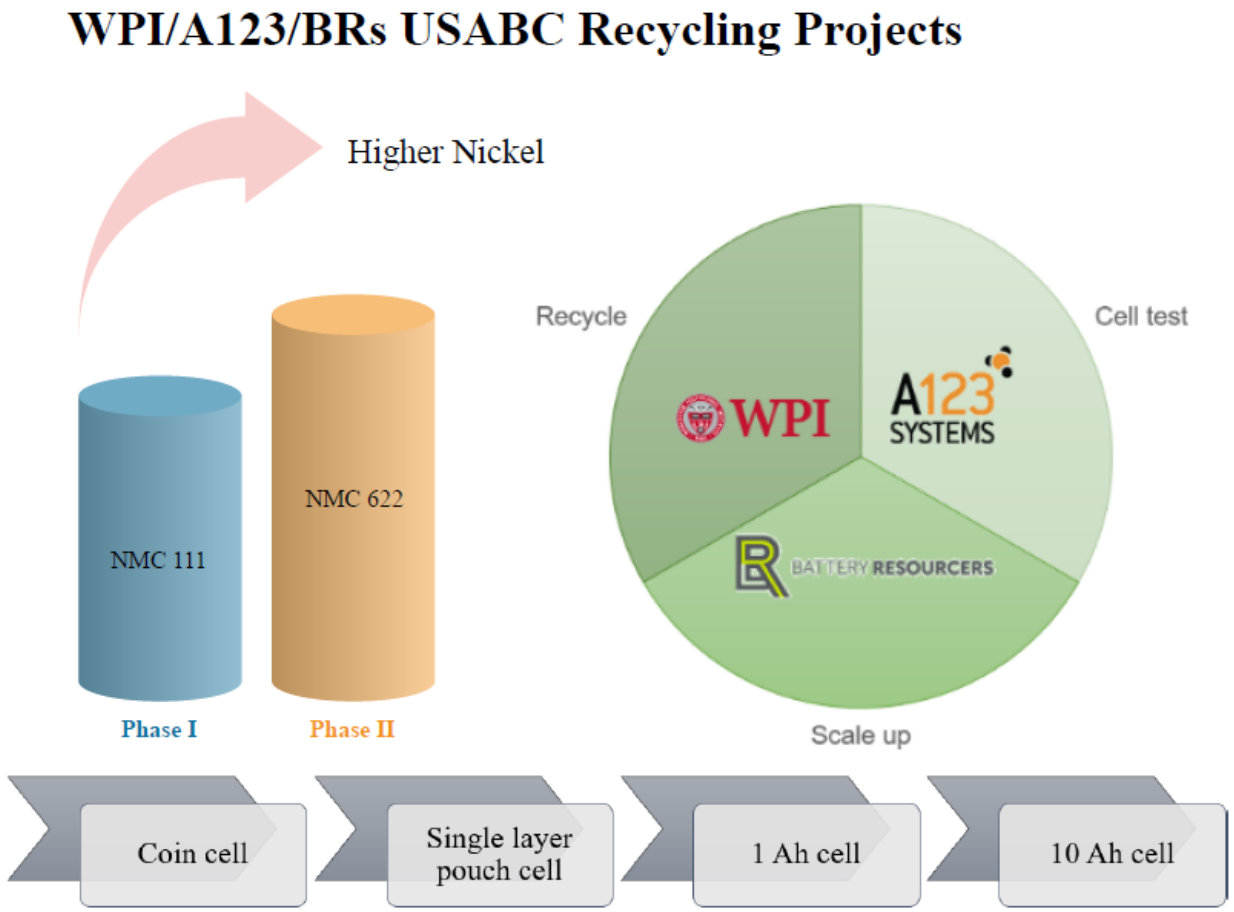


Build	Test Article	Total Made	USABC/ ANL	A123/ WPI	Kg powder per group
1	1 Ah NMC622/gr	20	12	8	6 (WPI)
1-control	1 Ah NMC622/gr	20	12	8	6 (comm.)
(2)*	(1 Ah NMC622/gr)*	(20)*	(12)*	(8)*	6 (WPI))*
3	10 Ah NMC622/gr	20	12	8	15
3-control	10 Ah NMC622/gr	20	12	8	15 (comm.)

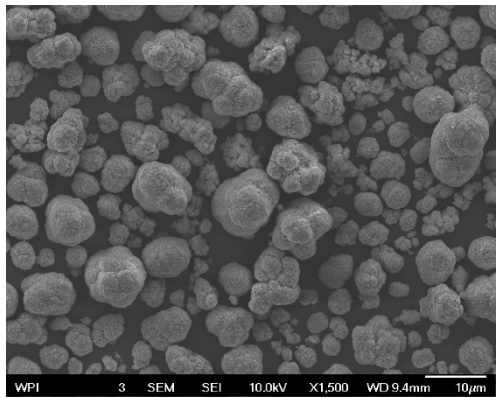
** 2nd 1 Ah build will only be executed if required. Depends upon 1st build results.*

Phase II: Approach/Strategy

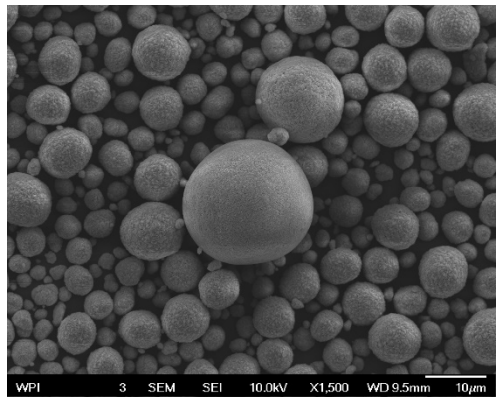
- Develop high nickel NMC from the spent EV batteries
- Understand the impacts of different anode materials and adhesives on the recycling process
- Develop scalable coating method for NMC622
- Recovered materials are tested in large pouch cells at A123 Systems
- The scale-up and cost model of the recycling process are developed at Battery Resourcers



Phase II: Technical Accomplishment and Progress



Day 1



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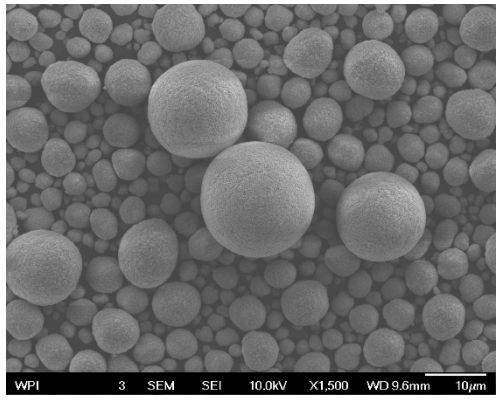

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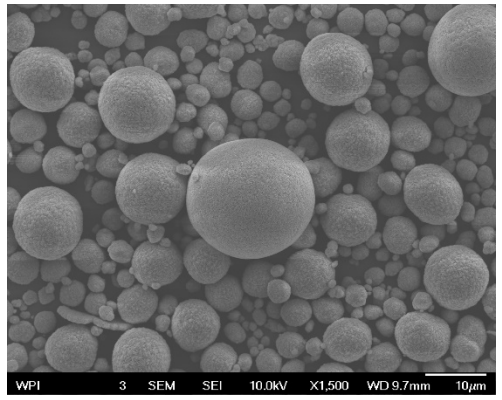

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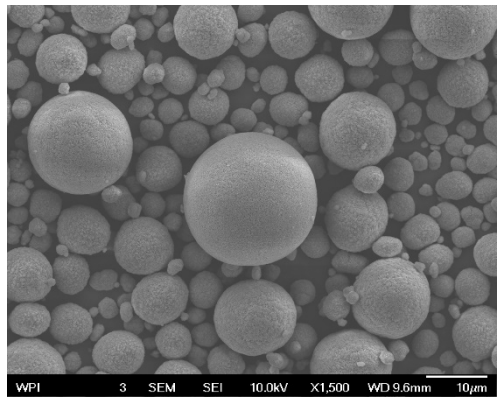

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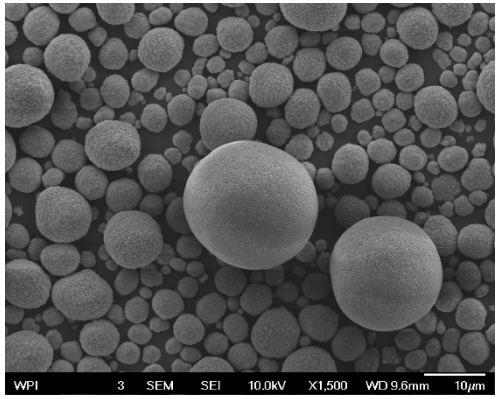

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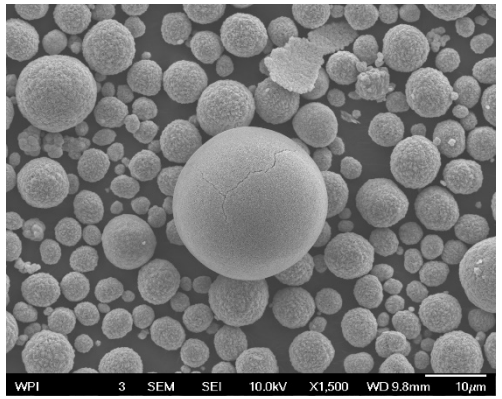

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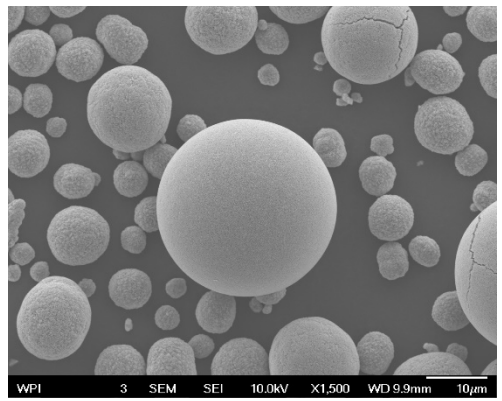

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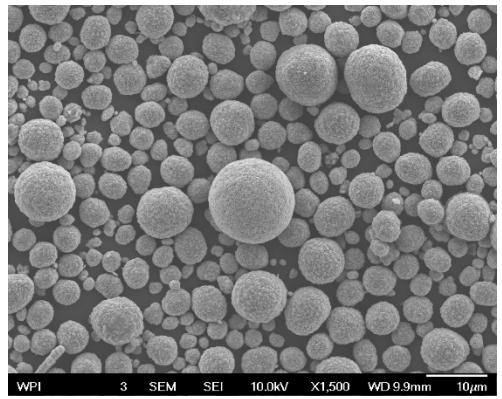

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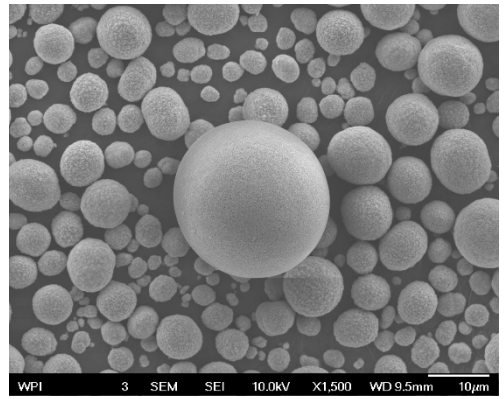
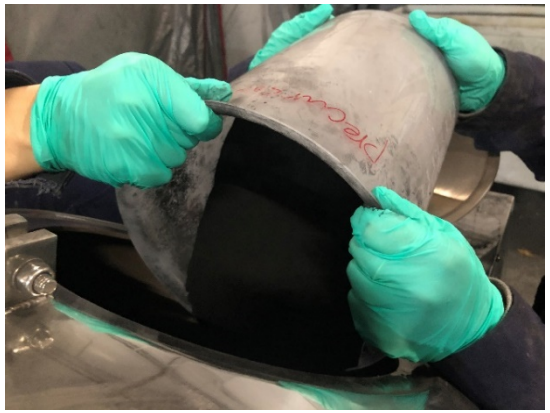


Day 27

NMC622 precursor with good morphology has been synthesized with the recycled materials from a continuous 27 days co-precipitation reaction.

Phase II: Cathode Sintering

Precursor



Precursor in V-mixer

Li2CO3

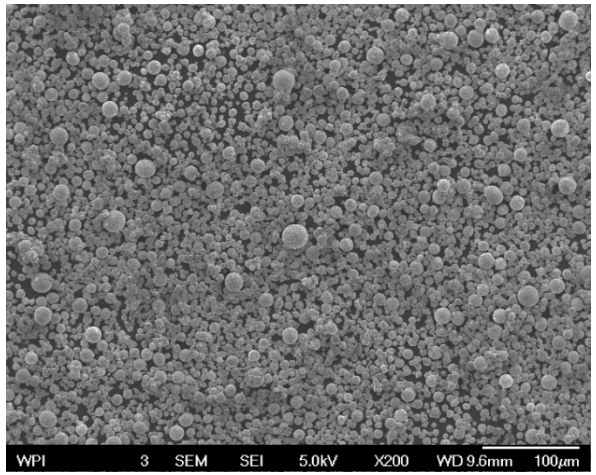
Mixing

Loading in Sagger

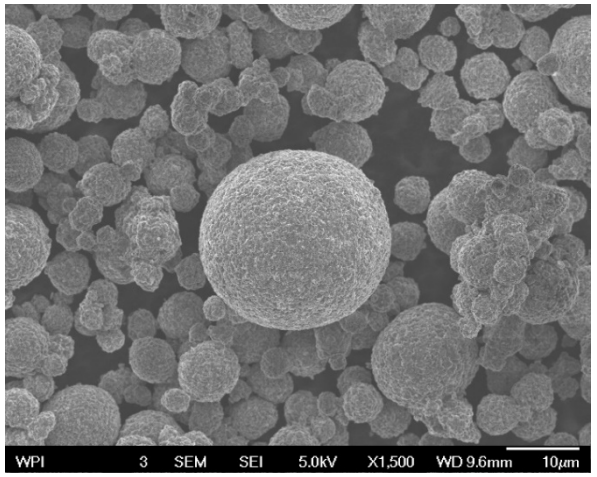
~20kg NMC622 cathode powder is sintered in Battery Resourcers pilot plant.

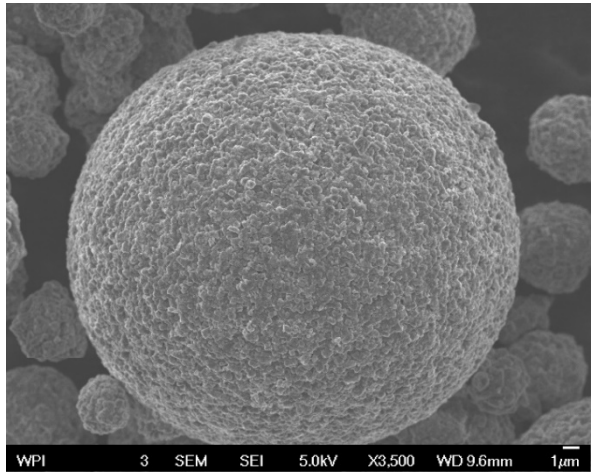
Phase II: Coated NMC622 Cathode Powder

x200

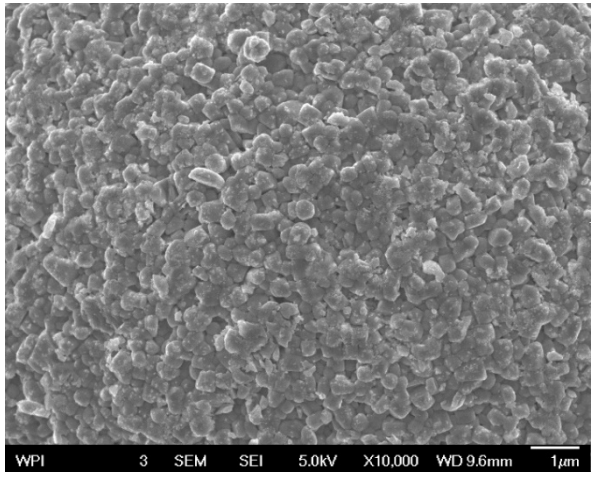


x1500



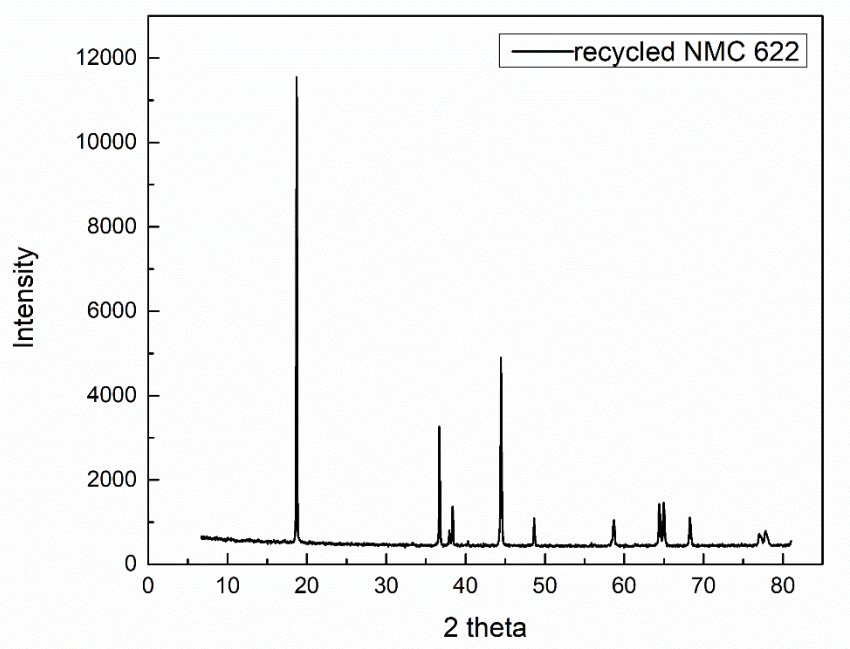


x3500



x10000

SEM shows spherical particles with bimodal distribution.

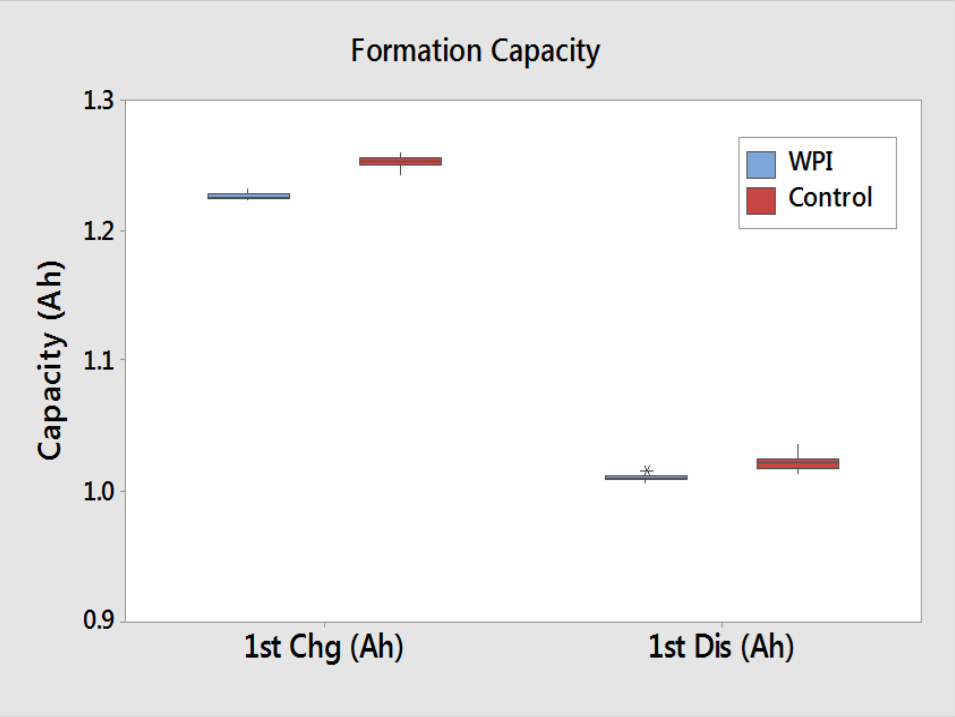


XRD shows typical layered structure.

Phase II: Coated Electrode at A123 Systems

Electrodes for 1Ah cells are coated with WPI Recovered cathode powder.

Phase II: A123 Test Results (1Ah Cells)



A123 Internal Testing Plan			
Test	Temperature	# of Cells	Current Status
Storage	60˚C	3	1month RPT done
Cycling 1C/1C	45˚C	3	Started 2/11/20
Rate Performance, then RT HPPC, then low temp HPPC	23˚C, 23˚C, 0˚C	3	Complete
On Hold	RT	3	30% SOC

Material	FCC (Ah)	FDC (Ah)	3rd discharge Cap (AH, C/2)	N-ICL (%) *(FCC-Retained Cap)/FCC*	ACR (mΩ)	Cell Thickness (mm)	Cell Weight (g)	Total # of cells built	# of cells shipped to ANL	# of cells tested at A123
WPI (TXS10443)	1.227	1.010	0.968	17.67	19.294	4.50	21.256	24	12	9
Control (TXS11158)	1.253	1.022	0.981	18.13	19.558	4.54	21.231	30	12	9

1Ah cells with WPI recovered NMC622 show comparable results with control powder.

Phase II: A123 Test Results (1Ah Cells)

Test Request	Cell #	Cycle 1 0.1C	Cycle 2 0.2C	Cycle 3 0.33C	Cycle 4 0.5C	Cycle 5 1C	Cycle 6 2C	Cycle 7 2.5C
TXS10443	4	100.00	98.99	97.90	96.80	94.31	88.13	75.69
TXS10443	5	100.00	98.92	97.61	96.58	94.04	88.46	77.60
TXS10443	6	100.00	98.93	97.82	96.67	94.05	87.20	74.26
Average (TXS10443)		**100.00**	**98.95**	**97.78**	**96.68**	**94.13**	**87.93**	**75.85**
TXS11158	14	100.00	98.46	96.93	95.53	92.61	87.44	79.49
TXS11158	18	100.00	98.55	97.04	95.61	92.72	87.37	79.20
TXS11158	21	100.00	98.50	96.93	95.56	92.71	87.79	80.90
Average (TXS11158)		**100.00**	**98.50**	**96.97**	**95.57**	**92.68**	**87.53**	**79.86**

+C/10, - xx/C
Retained Discharge Capacity Based on 0.1C Discharge Capacity (Values in %)
TXS10443 = WPI
TXS11158 = Control

1Ah cells with WPI recovered powder shows higher rate performance, except 2.5C.

Phase II: Recovered NMC622 for 11Ah Cells

PSD - WPI 02212020									
	batch 1	***batch 2***	***batch 3***	***batch 4***	***batch 5***	***batch 6***	***batch 7***	***batch 8***	***Total***
Dx (10)	5.39	5.31	5.95	6.09	6.17	5.98	6.59	6.18	
Dx (50)	9.99	9.88	11.7	11.8	11.7	11.4	11.9	12.2	
Dx (90)	18.3	18.7	22.4	21.7	21.2	21	21.2	23.3	
Dx (99.9)	29.7	33.1	37.2	34.6	34.5	34.6	34.7	39.2	
Amount (kg)	2.08	1.84	2.08	2.3	2.02	2.1	2.36	0.9	15.68

Echem			
	FCC (mAh/g)	FDC (mAh/g)	FCE (%)
WPI 02212020-3 *(11Ah build)*	195.0	172.7	88.5
WPI 08192019 *(1Ah build)*	194.2	175.2	90.2
Control	196.0	175.5	89.6

The delivered powder meets the requirements of particle size and electrochemical performance.

Scale-up and Commercialization

Based on the cost model developed at Battery Resourcers, the recycled cathode materials can be ~25% cheaper than the virgin materials.

Battery Resourcers is scaling up and commercializing the recycling process developed at WPI.

Responses to Previous Year Reviewers' Comments

Comment 1: The reviewer stated that the project is directly focused on materials supply and indirectly on sustainability of battery materials. The performance of the batteries produced from recycled materials is also addressed. The still-open question is if the secondary raw materials will be less expensive than the primary. The economic aspects are not presented by the project team yet.

Response: Based on the cost model developed at Battery Resourcers, the recovered cathode material is ~25% cheaper than the virgin materials. However, detailed information can not be shared.

Comment 2: The reviewer explained that the approach is an extension of the approach used during Phase 1 with NMC-111 recycled material. The data being generated with this approach can be used in general to make reasonable evaluations of the recycling process in terms of performance. However, it was not clear to the reviewer from the work presented how much variability in feedstock can be successfully handled with this specific recycling process. While the results are impressive so far on NMC-622 materials (similar to the earlier results on NMC- 111), the approach would be better if it clearly included a wide range of feedstock materials in terms of chemistry, binders, anodes, previous history, etc. The reviewer suggested that the approach should include enough variability in feedstock to be able to figure out the processing window—that is, how far from "typical material" can one go before the recycling process fails to produce satisfactory material?

Response: When planning the project, the team tried to incorporate all the possible materials from recycling streams. By working directly with GM, Ford, FCA and A123, we have secured cells with different cathode chemistry, anode chemistry and adhesives. Specially, the feedstock includes different cathode materials (NMC111, LMO, NCA and LFP), anode materials (graphite, LTO and Si) and adhesives from different OEMs.

Comment 3: The reviewer stated that it appears that good collaboration exists between A123, WPI, and battery resources, but added that for other collaborators, it appears less easy to judge the level of collaboration.

Response: The developers include WPI, A123 and Battery Resourcers, which did the main research work. Since this project is funded by USABC, the cells need to be independently tested by Argonne National Lab. The spent EV batteries are supplied by GM, Ford and FCA. SNT helps disassemble the entire battery packs to cells.

Collaboration and Partners

Scale-up and commercialization and cost analysis

Evaluate the recovered materials and fabricate commercial cells

Disassemble EV battery packs to cell level for recycling

Evaluate cells fabricated with recycled materials and virgin materials independantly

Go Further

Provide different battery packs for recycling experiments

Remaining Challenges and Barriers

- Understand the long term performance of the recycled NMC622 through 1Ah and 11Ah cells

Proposed Future Work

- Test 1Ah cells to determine the performance of both recycled and virgin materials
- Fabricate and test 11Ah cells to compare the performance of recycled and virgin materials
- Finish the cost model to show the economical benefits with recycling materials
- Consider to recover future cathode chemistry (for example NMC811 or above) and further lower the cost of recycled materials

Summary

- High performance NMC622 has been recovered from spent lithium ion batteries
- A continuous 27 days co-precipitation was successfully conducted
- 15kg cathode powder was delivered to A123 for 11 Ah cell fabrication
- 1Ah cells with recovered powder shows similar performance with control powder